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Modeling Non-Precipitating Cumulus Clouds as Flow-through-Reactor Transformer and Venting Transporter of Mixed Layer Pollutants

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16. ABSTRACT

A simple diagnostic model of cumulus convective clouds is developed and used in a sensitivity study to examine the extent to which the rate of change of mixed and cloud layer pollutant concentration is influenced by vertical transport and chemical transformation processes occurring as a result of cumulus clouds. The aqueous phase chemical transformation process is parameterized and modeled as a flow through batch reactor. The net vertical exchange between the mixed and cloud layers by cumulus cloud elements that protrude above the dry convective mixed layer is modeled as the product of a venting velocity and the difference in pollutant concentration between the mixed and cloud layer. In turn, the venting velocity is parameterized as proportional to the product of cloud cover and an effective cloud updraft. The two layer model yields an analytic expression involving key parameters such as areal extent of cloud cover, dry deposition rates, thickness in mixed and cloud layers, and the initial concentration difference between the mixed and cloud layers. The model solution was programmed using LOTUS 123.

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MODELING NON-PRECIPITATING CUMULUS CLOUDS AS FLOW-THROUGH-REACTOR TRANSFORMER AND VENTING TRANSPORTER OF MIXED LAYER POLLUTANTS

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Introduction:

Non-precipitating cumulus convective clouds can protrude well above the top of the convective mixed layer, as defined by the base of the elevated temperature inversion. These vigorous elements can, in principle, exchange momentum, moisture, and heat between the mixed layer and the overlying free troposphere and therefore, are important considerations in meso to large scale dynamic meteorological models (Anthes, 1985). These clouds also play an important role in the vertical transport and transformation of mixed layer pollutants. Experiments support the hypothesis that cumulus clouds do transport pollutants from the mixed layer to the overlying free troposphere (Ching et al., 1983, 1984, 1986; Greenhut et al., 1984 and Greenhut, 1986). Additionally, observational evidence clearly shows that aqueous phase chemical transformation rates within clouds are substantially greater than gas phase transformation rates of such pollutants as SO_2, and NO_x (Hegg and Hobbs, 1982).

Cumulus cloud fields exhibit great spatial, diurnal, seasonal, and geographical variability. It is therefore important to have an understanding of the extent to which such clouds influence pollutant concentration fields on regional scales since oxidant (Lamb 1983, and acid deposition (Chang, 1985) models on such scales now incorporate cloud processes in their development. Ching (1986) presented a highly parameterized and simplified diagnostic model to examine the role of cumulus clouds as vertical transporters of pollutants. In that study, clouds were classified as either active, passive, or forced as described in Stull (1985). Active clouds extend above the top of the mixed layer, passive clouds are remnants of active clouds, and fair weather cumulus are forced type clouds with vertical extent limited to the mixed layer. The net exchange by the active clouds is termed cloud venting (Ching, 1982). The diagnostic model predicted significant temporal changes in the mixed and cloud layer concentrations of a relatively conservative trace species such as ozone for a typical range of cloud venting activity. The purpose of this paper will be to extend the model by incorporating the effect of aqueous phase in-cloud pollutant transformation processes on the resultant mixed and cloud layer concentration fields.

The parameterization of the vertical exchange used by Ching (1986) is briefly summarized. The mass within active clouds acting over the lifetime of such clouds must remain aloft for net transport to occur. Cloud downdrafts induced by such clouds (Knupp and Cotton, 1985) are believed to play an important role in the net vertical exchange process (Ching and Alkezweeny, 1986). The manner in which downward motion induced by these upward venting clouds is viewed and the method by which it is parameterized in models is of critical importance to the predicted exchange rates. Updrafts and subsequent downdrafts induced by venting clouds can be envisioned to occur between two extremes. In one extreme, the

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penetrative updrafts are viewed as a temporary state followed by subsequent downward motion which returns the cloud volume (chemically converted) back into the mixed layer; as a result, little or no net vertical transport occurs. In the other extreme, maximum vertical exchange occurs when active cloud updrafts force free tropospheric air into the mixed layer concurrently as or soon after they rise above the mixed layer. Aircraft transects through such clouds (Greenhut, 1986) suggest that about 75% of the mixed layer mass within the active cloud elements will remain aloft, and ultimately become clouds of the passive type.

The diagnostic model presented here will examine the combined role of venting and in-cloud transformation on rates of change of pollutant concentration both in the mixed layer and in the free troposphere. This analysis is based on a coupled two-layer (mixed and free tropospheric) conservation of pollutant species model which distinguishes the role of active or venting type clouds from the forced, or fair weather cumulus clouds. The heterogeneous transformation processes are modeled as a flow-through-reactor system as applied to the forced type clouds.

Model Formulation:

The rate of change of concentration of a conservative pollutant in a grid volume bounded between the surface and the top of the mixed layer depends on the differential advection of the pollutant, the emission rate, the net gaseous and aqueous phase production or destruction, the net deposition to the surface, and the net flux of the pollutant out of the layer by cloud activity and is expressed in Equation 1 below:

$$\int_0^h \partial X/\partial t \, dz = \int_0^h (Adv+Em+P_g)dz + \int_0^h P_a dz - F_2 + F_1 \qquad (1)$$

Here, X = mixed layer pollutant concentration (ppb), Adv = advection component, Em is the emissions of pollutant, P_g is the gas phase pollutant production, P_a is the aqueous phase in-cloud pollutant production, F_2 = pollutant mass flux out of the mixed layer at h due to cloud venting and F_1 = deposition loss at surface. The surface deposition rate is typically parameterized as the product of a deposition velocity for the species times its concentration value. The terms P_g, Em and Adv will subsequently be treated together as a net source term, S.

Equation 2 refers to the pollutant concentration, Y, in the free troposphere or cloud layer whose vertical extent is the top of the mixed layer to the Limit of Free Convection, LFC, where the advection and source terms are neglected.

$$\int_h^{LFC} \partial Y/\partial t \, dz = F_2 \qquad (2)$$

The formulation for computing the cloud flux is from Ching (1986). The parameterization consists of (a), the difference of concentration between the two layers and (b), a scaling velocity for transport (ie., a "venting velocity", V_t) made up of fractional coverage of active clouds, α_v, whose updraft strength is w_c,

$$F_2 = \beta \, \alpha_v \, w_c \, (X-Y) = V_t \, (X-Y) \qquad (3)$$

where β is the coefficient that expresses the net effect of up and down drafts in determining the vertical exchange rate. Clearly, information on β, α_v, and w_c is needed to determine the venting velocity. The value of β used here is 0.74 from Greenhut (1986).

The cloud flux formulation depends explicitly on the concentration of both the mixed and the cloud layer. These expressions are greatly simplified and require the concentration within both the mixed layer and in the cloud layer to be instantly homogenized within their respective layers. Further, the details of the entrainment processes by dry convective motions at the top of the mixed layer are ignored. Thus, (1) and (2), after integration and rearrangement become:

$$\partial X/\partial t = S + P_a - (V_d + V_t)X/h + V_t Y/h \quad (4)$$

and

$$\partial Y/\partial t = V_t(X-Y)/\lambda \quad (5)$$

where λ is the thickness of the cloud layer, LFC-h.

The term, P_a, is parameterized in the following manner. Assume that forced clouds are formed by rising thermals with an updraft speed, w_f, that reaches its level of condensation (LCL) but does not protrude above h. Such updrafts contain within them, the pollutants from the emission layer. These pollutants flow through the aqueous, cloud environment, and finally, at some point in this movement, the cloud dissipates by entrainment or evaporates into the surroundings or becomes part of the return downdraft circulation thereby dropping below its LCL. In this manner the initial pollutant burden is rapidly transformed while in the aqueous in-cloud phase. This flow-through-reactor concept can be expressed as:

$$P_a = -\varepsilon \alpha_f w_f H X/h^2 \quad (6a)$$

$$\varepsilon = 1 - X_f/X \quad (6b)$$

where ε is the efficiency with which the in-cloud transformation process converts pre-cloud primary pollutant concentration, X, to a different species, X_f, and α_f is fractional area of forced clouds. For example, in the case of $X = SO_2$, X_f may be sulfate. Typically, X_f/X is less than 1 for primary pollutants, thus P_a is generally a sink term. Conversely, for secondary pollutants, Pa is usually greater than 1.

Expressions 6a,b are similar to Stull (1984), with H/h factored in to reflect the fact that forced clouds occupy only a fractional volume of the mixed layer due to its finite thickness H = h-LCL. Thus:

$$DX = S - V_f X - GX + F(Y-X) \quad (7)$$

and

$$DY = (h/\lambda) F (X-Y) \quad (8)$$

where $D = \partial / \partial t$

$G = V_d/h$

$F = V_t/h$

and $V_f = \varepsilon \alpha_f w_f H/h^2 = \varepsilon \alpha_f w_* H/h^2$

is the flow-through-reactor rate parameter. Observational evidence suggest substituting w_*, the convective scaling velocity, for w_f. Equation (7) is identical to (5) of Ching (1986), with the addition of the term $V_f X$. Substituting $J = V_f + G$

we obtain the identical form of the system of equations found in Ching (1986), with J replacing G. An interpretation of the term, J, is that the flow-through-reactor process exerts an influence on the rate of change of mixed layer concentration, X, in a manner effectively similar to the loss by surface deposition for ε greater than zero. On the other hand, when ε is negative, the chemical reaction increases the mixed layer pollutant species concentration. Setting the coefficients to constants, (7) and (8) can be readily solved and yield exact solutions below:

$$X = A(e^{m_1 t} - e^{m_2 t}) + B\, e^{m_2 t} + E \tag{9}$$

and

$$FY = A(m_1 e^{m_1 t} - m_2 e^{m_2 t} + (F+J)\,(e^{m_1 t} - e^{m_2 t}) + L\, e^{m_2 t} + KE \tag{10}$$

where

$$A = (FY_0 + S(m_2/J + 1) - X_0(m_2+F+J)/(m_1-m_2) \tag{11a}$$
$$B = X_0 - E \tag{11b}$$
$$E = S/J \tag{11c}$$
$$L = B\,(m_2+F+J) \tag{11d}$$
$$K = FE \tag{11e}$$
$$b = 0.5(J+F(1+h/\lambda)) \tag{11f}$$
$$m_1 = -b-0.5\,((b^2-4hFJ/\lambda))^{1/2} \tag{11g}$$
$$m_2 = -b+0.5\,((b^2-4hFJ/\lambda))^{1/2} \tag{11h}$$

X_0 and Y_0 are the initial concentrations. The requirement that the parameters and the source term be time invariant is an over simplification. The cloud convection time period is only likely to occur between late morning and late afternoon, when the cloud activity, deposition, production, mixed layer height and top of the cloud layer may exhibit some degree of stationarity.

Results and Discussion:

The major parameters for this two layer model are S, G, V_f, V_t, X_0, Y_0, h, and λ. In turn, V_f depends on ε, w_f (or its equivalent w_*), α_f, h and H. ε, of course, depends on the pollutant under consideration. It is, for example, expected to range from -0.5 for SO_4, near zero for ozone, and 0.5 for SO_2 and NO_x.

It is important to isolate the cloud processes using this model. If the term, S, representing the net effect of emissions, gas phase conversion, and advection balances the aqueous conversion and dry deposition losses, then the role of cloud venting can be illustrated more clearly. In this case, it can be shown that

$$W = W_0\, e^{-(2V_t/h)t} \tag{12}$$

where

$$W = X-Y$$

Thus, for $V_t = 0$, the concentration in both the mixed and cloud layer remain unchanged in time. However, for $V_t = 1$, 5, or 10 cm s^{-1}, the initial concentration difference between the mixed layer has dropped to 43%, 12% and 1% of its initial concentration difference respectively, after 6 hours.

Figure 1 shows the time changes in mixed layer and cloud layer ozone concentration for the just described situation where S balances both the surface deposition and the aqueous phase chemistry, and for which the venting velocity, V_t, was 10 cm s^{-1}. The other parameters are h= λ =1 km, the deposition velocity was assumed to be 1 cm s^{-1}, X_o = 50 ppb, Y_o = 0 ppb. To illustrate, V_t = 10 cm s^{-1} would apply to a cumulus updraft of 1 m s^{-1} for an areal coverage of 10% of cumulus clouds. Similarly, 2 m s^{-1} updrafts covering 5% yields V_t=10 cm s^{-1} etc. This example illustrates only the effect of cloud venting as a means of altering the concentrations in both the mixed and cloud layer. Within 6 hours, the concentrations between the mixed and cloud layers are nearly the same. Of course, this requires cloud activity to remain very large for the entire period of the simulation. The value of V_t used in this example is probably extremely large when considered over this period of time.

Figure 2 illustrates the case for which cloud venting is unimportant, and a special case when the term S (representing emissions, gas phase production, and advection) only balances the dry deposition surface loss term. Figure 2a, 2b, and 2c refer to fractional area of forced cumulus clouds α_f, of 1%, 10%, and 20%, respectively. This simulation might relate to SO_2 or NO_x pollutant, for which ε is assumed to be 0.5. Obviously, the greater the areal extent of forced cloud, the more rapidly the mixed layer concentration decreases with time. In this example, concentration decreased to 5%, 30%, and 50% of its initial concentration for α_f of 1%, 10%, and 20%, respectively, after 6 hours of flow-through-reactions.

Figure 3 shows the same situation as in Figure 2, but with the addition of venting cloud activity. In this case, V_t is lcm s^{-1}, a magnitude equivalent to the rate of dry deposition. It can be shown that this value of V_t is not large, but its impact sustained over the 6 hour simulation period is seen to be large. In contrast to Figure 2a, 2b, and 2c, the cloud layer pollutant concentration is shown to increase with time as was the case in Figure 1. The mixed layer pollutant concentration decreases 20%, 50% and 65% for α_f of 1%, 10%, and 20%, respectively, which are much greater decreases than in the non-venting cloud situation.

Finally, Figure 4a, 4b, and 4c refer to the case of very large venting velocity applied to the same case as for Figure 2. The venting velocity in this figure is 10 cm s^{-1}, typically a value too large to sustain over a duration of 6 hours (Cf discussion of Figure 1), but illustrates an extreme condition. The mixed layer concentration is seen to decrease to 45%, 55%, and 60% of its initial concentration for α_f of 1%, 10%, and 20% respectively. Due to the large magnitude of the venting effect in this example, the cloud layer concentration approaches the magnitude of the mixed layer concentration rapidly. The rate of pollutant homogenization between the two layer decrease from about 6 or so hours, to 2 hours and 1 hour for α_f of 1%, 10%, and 20% respectively.

These case situations do not refer to cloud coverage that are unreasonable. However, the resultant behavior of the rate of change of pollutant concentration in both layers are seen to be very sensitive to aqueous chemical conversion and to vertical transport processes. The range of cloud activity of both the forced and active cloud variety is clearly quite large in reality, and therefore, the treatment of these terms in regional scale models must be done with great care.

Summary:

This coupled mixed-cloud layer model is designed to diagnose the changes in pollutant concentration in both layers as a result of cumulus cloud activity. Processes incorporated in this model include; vertical exchange by cloud venting, dry deposition to the surface, aqueous phase chemistry in the fair weather cumulus clouds, gas phase production and advection. Coefficients related to deposition, venting, gas and aqueous phase transformation rates and advection are time invariants in this study. The formulation for the flux is the product of a venting velocity and the concentration difference between the mixed and cloud layers. The venting clouds are the active clouds defined by Stull (1985); his forced clouds are the fair weather cumulus which are restricted to the mixed layer; active clouds are not treated in detail in this study. The venting velocity parameterization incorporates cloud induced downdrafts implicitly in its formulation. The formulation of the venting velocity is based on empirical data collected from aircraft observations. The aqueous phase reactions are a sink for primary pollutants such as SO_2 and NO_x, and are believed to be efficient transformation processes and effective in reducing concentrations. Great variability is predicted due to aqueous phase chemistry because of the large spatial and temporal variability in α_f.

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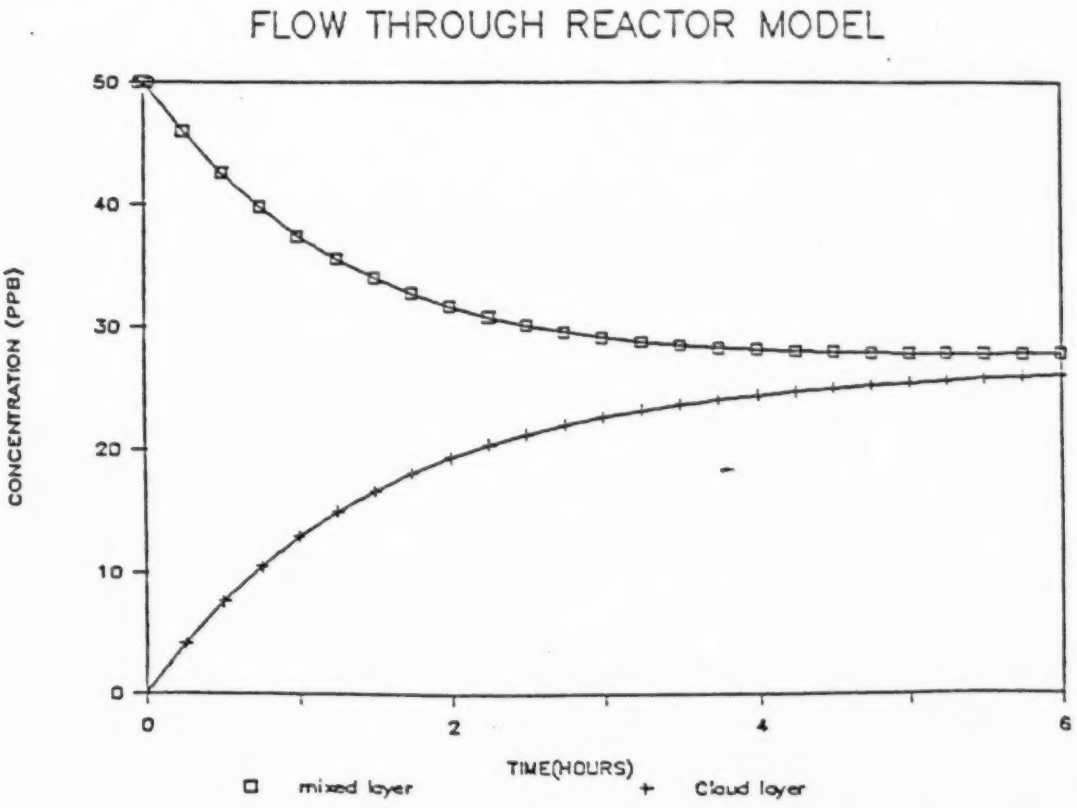

Figure 1. Modeled ozone concentrations in the mixed and cloud layers. This case assumes a balance between the net production term and the surface deposition. V_t = 10 cm s^{-1} throughout the simulation, X_o and Y_o were 50 and 0 ppb, respectively.

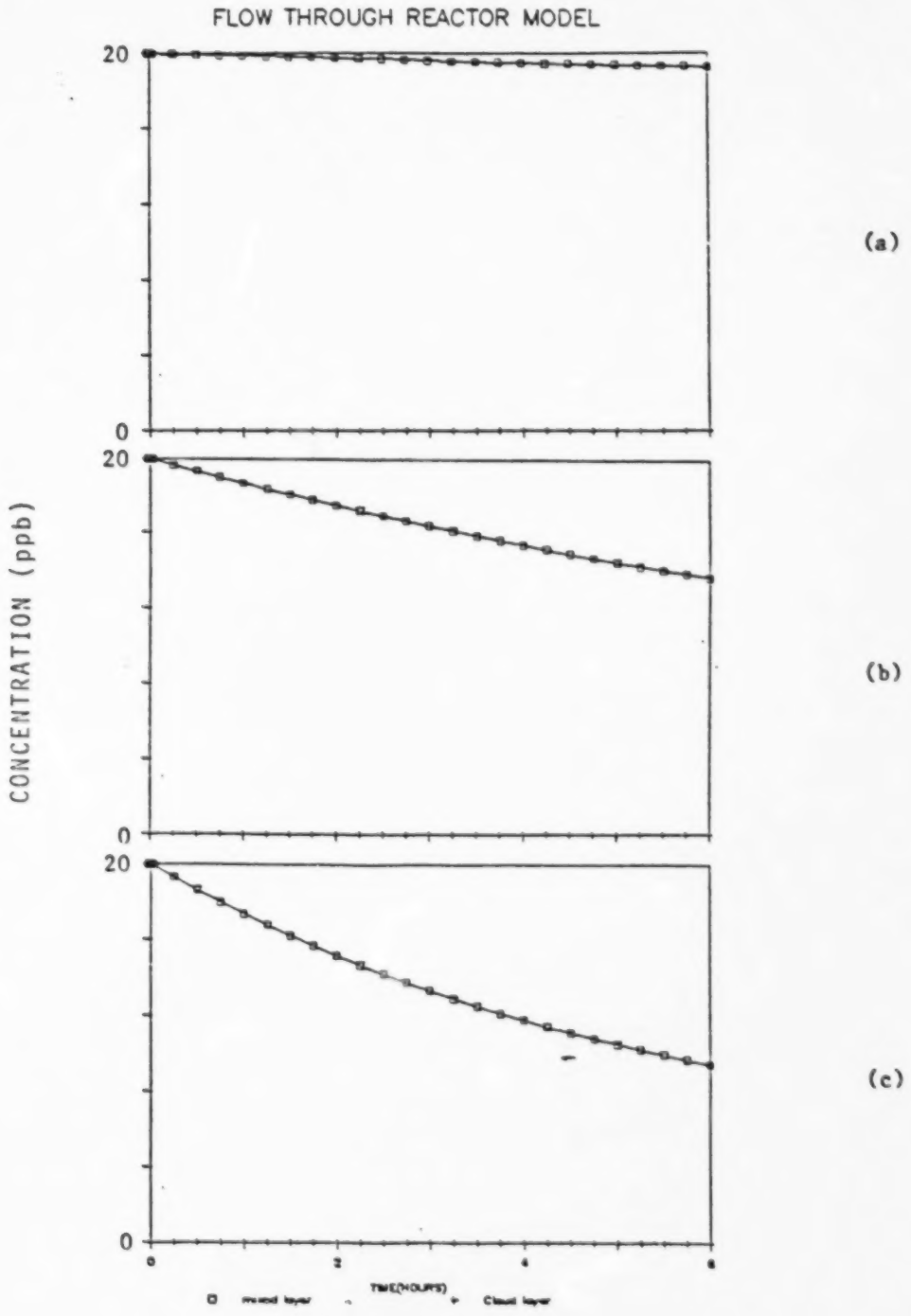

Figure 2. Modeled reactive pollutant concentrations in mixed laver. Concentrations in cloud layer unchanged because $V_t = 0$. The aqueous phase chemistry is shown for forced cloud activity of α = a) 1%, b) 10% and c) 20%, and X_o = 20 ppb.

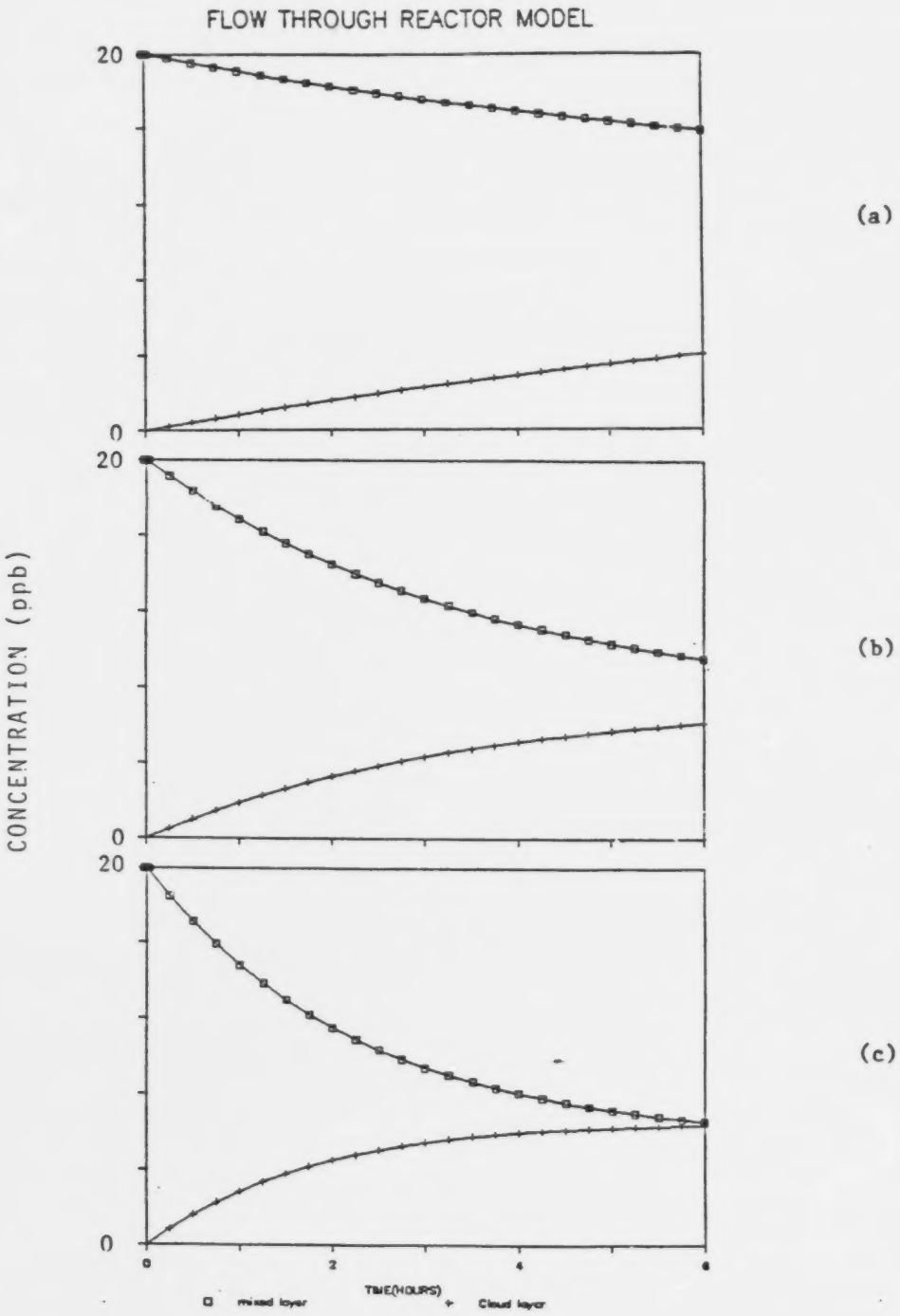

Figure 3. Same as Figure 2 except V_t = 1 cm s^{-1} applied over the simulation period and Y_o = 0 ppb.

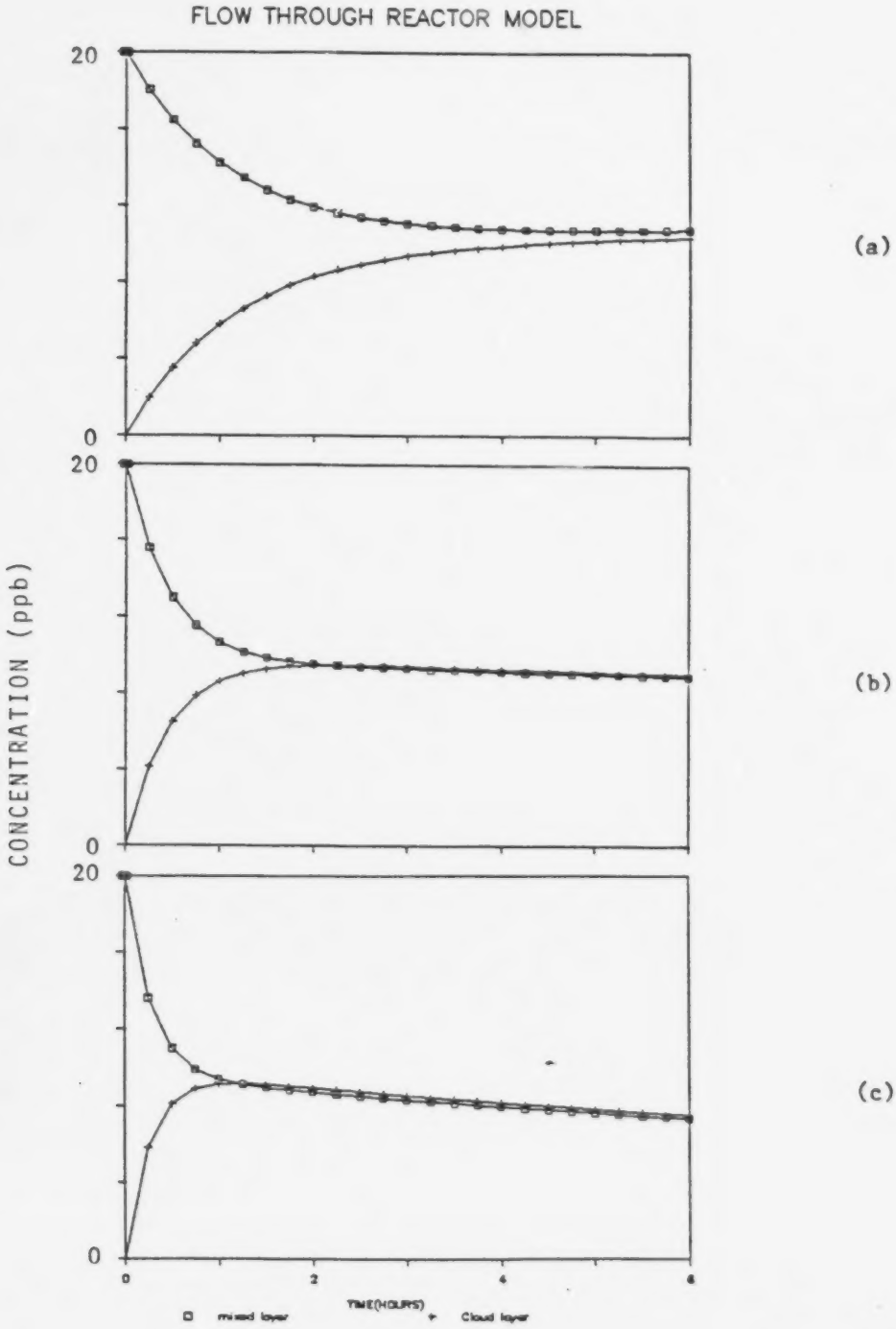

Figure 4. Same as Figure 3 except that V_t = 10 cm s^{-1}.